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(19) (CA) **APPLICATION FOR CANADIAN PATENT** (12)

(54) Composite Particles, Method for Preparing the Same and  
Amino Resin Composition Containing the Particles

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## Abstract of the Disclosure

Composite particles comprising talc particles covered with a layer of a condensate of formaldehyde and at least one member selected from the group consisting of aminotriazine type compounds, urea and phenols, the talc surface being chemically bonded to the condensate layer. The composite particles can be prepared by a method which comprises reacting formaldehyde with at least one member selected from the group consisting of aminotriazine type compounds, urea and phenols in an aqueous medium in the presence of talc particles to form a layer of a condensate on the surface of the talc particles. Amino resin compositions containing the composite particles have mechanical strength comparable to those containing a cellulosic filler and excellent resistance to thermal discoloration and, therefore, they can be used as molding materials for heat resistant electric machine parts and heat resistant tablewares.

## SPECIFICATION

## TITLE OF THE INVENTION

Composite Particles, Method for Preparing the Same and  
Amino Resin Composition Containing the Particles

## 5 SUMMARY OF THE INVENTION

(Field of the Invention)

The present invention relates to composite particles useful as a  
filler for thermoplastic and thermosetting resins (hereinafter  
referred to as "plastics"), a method for preparing the particles and a  
10 resin composition containing the particles.

(Description of the Prior Art)

Fine powdery talc is an inorganic filler which has been used in  
great quantities as a filler for plastics, paper making or the like.  
If an inorganic filler is incorporated into a plastic, the elastic  
15 modulus and heat deformation temperature of the plastic are generally  
improved, but the tensile strength and impact strength thereof are  
often lowered. It is considered that this is due to insufficient  
affinity of a plastic for an inorganic filler. For this reason,  
various methods have been investigated to enhance the affinity of the  
20 surface of an inorganic filler for a plastic. For instance, there have  
been known a method in which a variety of coupling agents such as  
silane type, titanium type or chromium type ones; and a method using  
a surface treating agent such as fats and oils, a higher fatty acid, a  
higher alcohol, a plasticizer or a high boiling point hydrocarbon.

25 However, it has been known that, in most of cases, a desired  
effect due to the addition of a coupling agent or a surface treating

agent is not attained for talc and it is very difficult to incorporate a large amount of talc into some plastics without impairing mechanical properties of the plastics. It is believed that the physical properties of a composite plastic are not improved sufficiently by simply improving the affinity of the surface thereof for a plastic. This is because the foregoing methods are not sufficient for improving the affinity of talc for a plastic, the crystalline structure of talc is layer structure and it is liable to cause the ply separation.

On the other hand, amino resin molding materials have various excellent properties such as resistance to arc, tracking resistance, electrical insulation properties, burning resistance, coloring properties, surface hardness, solvent resistance and mechanical properties compared with other plastics, but they are inferior in resistance to thermal discoloration and, therefore, they are limited in their applications. It is known that the thermal discoloration is mainly due to the presence of cellulose type fillers commonly used. To solve this problem, the use of an inorganic filler is desired.

As such inorganic fillers, there have been used, for a long time, glass fibers and asbestos. As other examples of inorganic fillers used in composite plastics, Japanese Patent Un-examined Publication (hereunder referred to as "J.P. KOKAI") No. Sho 57-67650 discloses a melamine resin composition as a molding material in which  $\beta$ -type calcium metasilicate and talc are simultaneously incorporated; and J.P. KOKAI No. Sho 53-51242 discloses an amino type resin composition as a molding material in which aluminum hydroxide and talc are incorporated.

However, such conventional inorganic fillers suffer from the following problems and it has long been desired to solve the problems.

For instance, in the case of fibrous fillers such as glass fibers and asbestos, the warp of a molded article is increased due to orientation of the fibers, and asbestos possibly have carcinogenicity. On the other hand, in the case of talc,  $\beta$ -type calcium metasilicate, calcium carbonate and aluminum hydroxide having a relatively low aspect ratio, it is impossible to impart sufficient mechanical strength to the resultant composite plastics.

#### SUMMARY OF THE INVENTION

Accordingly, a primary object of the present invention is to provide composite particles useful as a filler for plastic composite materials. The particles are composed of talc particles whose surface is covered with a layer of an organic material having sufficient mechanical strength and a proper thickness.

Another object of the present invention is to provide a method for preparing such composite particles.

A further object of the present invention is to provide an amino resin composition containing the above defined composite particles. The resin composition is excellent in resistance to thermo-discoloration and exhibits mechanical strength comparable to that of the resin composition containing a cellulosic filler.

The inventors of this invention consider that if the surface of talc particles are covered with a layer of an organic material having sufficient mechanical strength and a proper thickness, the affinity thereof for plastics and the tendency of the ply separation of talc

crystals would be improved and, as a result, the physical properties of a plastic composite material into which such composite particles are incorporated would be improved and have conducted various studies to solve the foregoing problems. Consequently, the inventors have  
5 found that the foregoing objects of this invention can effectively be achieved by using composite particles obtained by reacting, in the presence of talc particles, formaldehyde with at least one member selected from the group consisting of aminotriazine compounds, urea and phenols in an aqueous medium to thus form a layer of thermosetting  
10 resin condensate on the surface of the talc particles and thus have completed the present invention.

According to an aspect of the present invention, there is provided composite particles which comprises talc particles covered with a layer of a condensate of at least one member selected from the  
15 group consisting of formaldehyde and formaldehyde-generating compounds and at least one member selected from the group consisting of aminotriazine compounds, urea and phenols, the talc surface being chemically bonded to the condensate layer.

According to another aspect of the present invention, there is  
20 provided a method for preparing such composite particles of talc which comprises reacting, in the presence of talc particles, at least one member selected from the group consisting of formaldehyde and formaldehyde-containing compounds with at least one member selected from the group consisting of aminotriazine compounds, urea and phenols  
25 in an aqueous medium to thus form a layer of thermosetting resin condensate on the surface of the talc particles.

According to a further aspect of the present invention, there is provided an amino resin composition comprising the above defined composite particles.

#### DETAILED EXPLANATION OF THE INVENTION

5           The present invention will hereinafter be explained in more detail.

          The talc used in the invention is gray to white colored inorganic powder obtained by pulverizing an ore, talc (which is chemically magnesium silicate hydrate represented by  $3\text{MgO} \cdot 4\text{SiO}_2 \cdot \text{H}_2\text{O}$ ) and preferably those commercially available having a particle  
10           size of not more than 60 mesh.

          Examples of the aminotriazine type compounds used herein are melamine and guanamines such as benzoguanamine and acetoguanamine. Regarding urea used in the invention, a part thereof may be replaced  
15           with thiourea. Examples of the phenols used herein include monovalent phenols such as phenol, cresol, xylenol, t-butylphenol, phenylphenol, naphthol, alkyl phenols whose alkyl group generally has 2 to 12 carbon atoms, cyclohexylphenol, halogen-substituted phenols, and bivalent phenols such as resorcin, bisphenol A, bis(hydroxyphenyl)  
20           methane, bis(hydroxyphenyl) ether, bisphenol B and bisphenol S. Examples of formaldehyde generating compounds include formalin, trioxane and paraformaldehyde. The term "formaldehyde" hereinafter means also such formaldehyde-generating compounds.

          In the method of this invention, two or more of the compounds  
25           selected from the group consisting of aminotriazine compounds, urea and phenols may also be used to coat the surface of the talc particles



with a co-condensed resin layer. For instance, co-condensed resins such as urea-melamine co-condensed resin and melamine-phenol co-condensed resin are also suitably used in the invention.

5 The term "condensate layer or layer of condensate" as used herein means those formed through a condensation reaction of formaldehyde with at least one member selected from the group consisting of aminotriazine type compounds, urea and phenols (hereinafter referred to as "amine · phenols"). The degree of condensation of the condensate layer may be in either "B-stage" or  
10 "C-stage" expressed by the term generally used for expressing the degree of condensation of thermosetting resins. The thickness of the condensate layer may vary depending on intended purposes of the composite particles.

The degree of condensation of the condensate layer can be  
15 increased during the preparation of the composite particles of the present invention or when the composite particles are incorporated into plastics or rubbers to thus obtain a composite plastic or a composite rubber, by a known method. Whereby the mechanical strength of the condensate layer reaches the level of usual thermosetting  
20 resins and thus a layer of an organic substance having satisfactory mechanical strength.

The chemical bond between the talc surface and the condensate layer will now be explained below. It is generally known that particles of talc and other silicate compounds in general have  
25 hydroxyl groups on the surface. As will be explained below, it is assumed that if formaldehyde is reacted with amine · phenols in the



presence of talc particles in an aqueous medium, the hydroxyl groups on the talc surface react with formaldehyde to form active sites such as methylol groups on the talc surface and the active sites serve as starting points for forming water-insoluble condensate layer on the talc surface. More specifically, the talc surface is chemically bonded to the condensate layer. The presence of such chemical bonds is proved by the following two facts: As will be described in, for instance, Example 1, all of the condensates at the end of the reaction are substantially water-insoluble if proper reaction conditions are selected; and if the resulting particles are observed under a scanning electron microscope, the composite particles of the present invention are exclusively formed and hence particles consisting of the thermosetting resin free of talc particles are not observed. From these facts, it can be concluded that substantially all of the condensates are used for covering the talc surface. It is well known to those skilled in the art that the condensation reaction proceeds, at least at the beginning of the reaction, mainly in an aqueous phase. However, the fact that substantially all of the condensates are attached to the talc particles can be explained only by the presence of chemical bonds between the talc surface and the condensates. It is considered that such chemical bonds may be formed by the following mechanism. More specifically, functional groups such as the methylol groups (or methylol ether groups) serve as active sites for the reaction and the active sites react with condensates formed in an aqueous phase at the initial stage of the reaction or the amine phenols to form new active sites. The active sites thus formed in turn

react with initially formed condensates in the aqueous phase to generate new active sites. The condensate layer gradually grows on the talc surface through the repetition of such reactions and finally the condensates in the aqueous phase are substantially exhausted.

5           Moreover, the fact that the amount of the condensate layer formed is greatly reduced if the reaction conditions are not proper, for instance, if water and talc particles are added to an initial condensation solution of formaldehyde and melamine and the mixture is heated and stirred for several hours, also demonstrates the idea that  
10           the condensate layer is not formed by a simple physical adsorption on the talc surface.

          In the composite particles of this invention, the condensate layer is strongly bonded to the talc surface through chemical bonds and, therefore, they essentially differ from those obtained by, for  
15           instance, a method comprising simply physically mixing talc particles and initial condensation solution of a thermosetting resin prepared in an ordinary manner and then drying; or a method comprising coating the surface of talc particles with a thermosetting resin melted by heating.

20           The content of the talc particles in the composite particles of this invention preferably ranges from 10 to 95% by weight. This is because if it is less than 10% by weight, the control of particle size becomes difficult, while if it exceeds 95% by weight, the properties of the composite particles as a filler is liable to be  
25           impaired.

          The composite particles of the present invention thus prepared

may undergo secondary aggregation of composite particles coated with the condensate layer by properly selecting the reaction conditions to thus form enlarged composite particles. The degree of the secondary aggregation can be controlled by properly adjusting the reaction conditions as will be detailed below and whereby the particles size can be controlled in a wide range of from the size of the composite particles substantially consisting of primary particles to the size of those comprising enlarged composite particles composed of several hundreds of primary particles.

The method for preparing the composite particles of this invention will now be explained below.

In the method of this invention, formaldehyde and amine · phenols are first reacted in an aqueous medium in the presence of talc particles.

The order of the addition of each component is not critical, but in general a desired amount of talc particles are first suspended in water, then formaldehyde is added thereto and finally the amine · phenols are added. The foregoing order of addition is desired since there is observed a tendency that the yield of the resulting solid contents is possibly high when formaldehyde is added prior to the addition of the amine · phenols.

The term "solid contents" herein means the sum of talc particles and the condensate layer coating the surface of the talc particles. The condensate layer is water-insoluble at the end of the foregoing reaction.

It is found that if the talc particles are treated with an acid

prior to the condensation reaction, the yield of the solid contents is liable to increase compared with that observed when the particles are not treated with an acid.

5 Such an acid treatment may be performed by previously bringing the particles into contact with an acid outside the reaction system or in the reaction system, more specifically by adding an acid so that pH of the system is not reduced to 2 or less, after suspending the talc particles in water. The kinds of acids used are not restricted to specific ones. Examples thereof used include inorganic  
10 acids such as hydrochloric acid, sulfuric acid, phosphoric acid and boric acid; carboxylic acids such as formic acid, oxalic acid, acetic acid, tartaric acid, citric acid and maleic acid; and sulfonic acids such as p-toluenesulfonic acid and alkylbenzenesulfonic acid whose alkyl group has 10 to 18 carbon atoms.

15 In the method of this invention, a basic catalyst can be used in the foregoing condensation reaction.

As such basic catalysts, there may be used those conventionally used for the preparation of melamine resin, urea resin or resol type phenol resins and specific examples thereof are aqueous ammonia,  
20 pyridine, hexamethylenetetramine, alkylamines such as dimethylamine, diethylamine and polyethyleneimine, and inorganic bases or inorganic salts such as sodium hydroxide, potassium hydroxide, sodium carbonate, potassium carbonate and calcium hydroxide.

The condensation reaction is performed in an aqueous medium. In  
25 this case, it is desired that the amount of the aqueous medium is selected so that the solid contents at the end of the reaction ranges

from 10 to 80% by weight and preferably 30 to 70% by weight. The condensation reaction in the method of this invention is desirably carried out at 50 to 100° C for 1 to 10 hours with stirring or kneading.

5           In the method of this invention, the degree of secondary aggregation in the condensate layer can be controlled by properly adjusting the reaction conditions such as the ratio of the added amount of talc particles and that of the amine · phenols, the kinds of the amine · phenols used, the reaction temperature, the reaction time, 10 speed of stirring or kneading, and the kinds or concentration of the surfactants used or the concentration of water-insoluble solids. The molar ratio of formaldehyde to the amine · phenols desirably falls within the range usually adopted to form melamine resin, urea resin, phenol resin, urea-melamine co-condensed resin or melamine-phenol co- 15 condensed resin and it generally ranges from 1 to 3. Moreover, the degree of condensation and the degree of cure must be a level at which the resin is water-insoluble at the end of the reaction due to the chemical bonds with the surface of the talc particles or the cross-linking reaction between the condensates. In addition, it is also possible to promote the cross-linking reaction of the condensates by 20 adding an acid at the time when the reaction proceeds to some extent. Examples of acids used may be the same as those defined above in connection with the acid treatment of the talc particles.

25           In the method of this invention, a surfactant may be used to control the degree of secondary aggregation of the composite particles as has been described above. The kinds of the surfactants is

not critical and various kinds of surfactants commercially available (cationic, anionic and non-ionic ones) may be used alone or in combination.

5 In the present invention, pulp may simultaneously be used. More specifically, the composite particles of the present invention may be prepared by reacting formaldehyde and the amine · phenols in the coexistence of the talc particles and pulp. The amount of the pulp is 0 to 90% by weight, preferably 0 to 50% by weight based on the total weight of the pulp and the talc particles.

10 After the completion of the reaction, the reaction system is cooled to a temperature of not more than 50° C, followed by optionally separating the resulting solids from liquid in an ordinary manner such as filtration or centrifugation and then drying to thus obtain the composite particles of this invention. Upon drying the  
15 resulting particles, the degree of condensation can be controlled to some degree by properly selecting the drying temperature and time.

The composite particles of the present invention prepared according to the method of this invention are covered with a layer of an organic material having sufficient mechanical strength and a proper  
20 thickness and the particle may have any particle size over a wide range.

According to another aspect of the present invention, there is provided an amino resin composition containing the foregoing composite particles.

25 Examples of the amino resins used in the invention are preferably melamine resins, urea resins, guanamine resins and their



modified resins such as phenol-melamine resins, guanamine-melamine resins, phenol-urea resins, urea-melamine resins, phenol-guanamine resins and urea-guanamine resins. In this respect, at least one member selected from the group consisting of benzoguanamine and acetoguanamine may be used as the guanamine component. In addition, at least one member selected from the group consisting of the foregoing phenols may be used as the phenol component.

The content of the composite particles in the amino resin composition of the present invention preferably ranges from 5 to 70% by weight on the basis of the total weight of the composition. This is because if it is less than 5% by weight, the effect achieved by the addition of such composite particles becomes insufficient, while if it exceeds 70% by weight, the moldability of the resulting composition is liable to be lowered.

Methods for preparing the amino resin composition of the present invention from the foregoing amino resin and the composite particles are not restricted to specific ones and various methods usually employed may be adopted. Examples thereof are a method comprising mixing the composite particles and an initial condensation solution of an amino resin and then drying the mixture; or a method comprising pulverizing, mixing or kneading a solid amino resin and the composite particles with a commonly used means such as a ball mill, a roll mill or a kneader. The amino resin composition of this invention may further comprise curing agents, releasing agents and coloring agents commonly utilized. It is also optionally possible to, simultaneously with the composite particles, incorporate at least one member selected



from the group consisting of cellulosic fillers such as pulp and wood powder and inorganic fillers such as calcium carbonate and aluminum hydroxide into the amino resin.

5       The composite particles of the present invention are covered with a layer of a condensate of formaldehyde and amine · phenols and the condensate layer is chemically bonded to the talc surface. Therefore, the composite particles have a true specific gravity smaller than that of talc particles per se; good affinity for or compatibility with organic substances; high mechanical strength; high  
10       heat resistance and resistance to thermal discoloration; as well as excellent flame resistance. By making most use of such characteristics, the composite particles can be effectively used as reinforcing agents, fillers for a variety of plastics, rubbers or the like.

15       It is optionally possible to obtain enlarged composite particles from the talc particles covered with the condensate layer by subjecting them to secondary aggregation.

In addition, the composite particles of the invention can very easily and stably be prepared by the method of this invention.

20       Moreover, the amino resin composition of the present invention is excellent in resistance to thermal discoloration, has mechanical strength identical with or greater than that of the composition containing a cellulosic filler and the molded articles obtained from the composition exhibit warp or bending in a very low extent. The  
25       amino resin composition of the present invention can be used as a material for heat resistant electric machine parts, heat resistant

tablewares or the like by making most use of these characteristics.

The present invention will hereunder be explained in more detail with reference to the following non-limitative working Examples and the effects practically achieved by the present invention will also be discussed in comparison with Comparative Examples.

5

#### Example 1

1200 g of water was charged in a 4ℓ separable flask provided with a cooling apparatus and 350 g of talc particles having a particle size of  $2.3\mu\text{m}$  was introduced into the flask with stirring to obtain a suspension of talc particles. To the suspension, there was added 2.5 g of 10% by weight hydrochloric acid solution and it was stirred at room temperature for 30 minutes. At this stage, pH of the suspension was 6.8 (BTB). The pH value was not lowered at all. This is due to the neutralization of acids by talc partially dissolved out. After adding 300 g of 37 wt% formalin, the temperature of the suspension was raised to  $90^{\circ}\text{C}$  over 40 minutes and this temperature was maintained for 20 minutes. Then 240 g of melamine was added to perform a reaction at  $90^{\circ}\text{C}$  for 5 hours with stirring.

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Then the contents of the flask was cooled to  $50^{\circ}\text{C}$  and the contents were washed with 2 ℓ of water while carrying out filtration. The resulting solid contents were dried at  $80^{\circ}\text{C}$  in a hot-air dryer till the weight of the solid became constant. The yield of the solid content was 638 g. Thus, the talc particles covered with a layer of the condensate of formaldehyde and melamine, i.e., the composite particles of this invention were obtained. The composite particles were observed under an electron microscope and found that the average

particle size thereof was  $20\mu\text{m}$ , that the secondary particles were formed and that there were not observed the presence of particles comprising only the thermosetting resin.

#### Example 2

5           1000 g of water was charged in a 4ℓ separable flask provided with a cooling apparatus and 300 g of talc particles having a particle size of  $2.3\mu\text{m}$  was introduced into the flask with stirring to obtain a suspension of talc. To the suspension, there was added 2.5 g of 10% by weight hydrochloric acid solution and it was stirred at room  
10           temperature for 30 minutes. After adding 360 g of 37 wt% formalin, the temperature of the suspension was raised to  $60^\circ\text{C}$  and this temperature was maintained for 40 minutes. Then 180 g of urea was added and the reaction was carried out at that temperature for 5 hours with gradually adding 80 g of 10wt% hydrochloric acid in several  
15           portions. Then sodium carbonate was added to adjust pH thereof to 7. Then the contents of the flask were cooled to  $50^\circ\text{C}$  and the contents were washed with 2 ℓ of water while carrying out filtration. The resulting solid contents were dried at  $80^\circ\text{C}$  in a hot-air dryer till the weight of the solid became constant. The yield of the solid  
20           contents was 503 g. Thus, the talc particles covered with a layer of the condensate of formaldehyde and urea, i.e., the composite particles of this invention were obtained. The composite particles had an average particle size of about  $3\mu\text{m}$  and there was not observed the presence of secondary particles.

#### 25           Example 3

          1400 g of water was charged in a 4ℓ separable flask provided

with a cooling apparatus and 400 g of talc particles having a particle size of  $2.3\mu\text{m}$  was introduced into the flask with stirring. After adding 100 g of 37 wt% formalin, the temperature of the suspension was raised and 100 g of phenol and 4 g of dry sodium carbonate were added to the suspension at the time when the temperature reached  $85^\circ\text{C}$ . The reaction was continued at that temperature for 3 hours and the reaction was continued for additional 2 hours after slowly adding 100 g of 10% hydrochloric acid in small portions. Then sodium carbonate was added to neutralize the reaction system to thus adjust pH of the reaction liquid to 7.

Then the contents of the flask were cooled to  $50^\circ\text{C}$  and the contents were washed with 2  $\ell$  of water while carrying out filtration. The resulting solid contents were dried at  $80^\circ\text{C}$  in a hot-air dryer till the weight of the solid became constant. The yield of the solid content was 490 g. Thus, the talc particles covered with a layer of the condensate of formaldehyde and phenol, i.e., the composite particles of this invention were obtained. The composite particles had an average particle size of about  $50\mu\text{m}$ .

#### Example 4

To a 4 $\ell$  separable flask provided with a cooling apparatus, there were added 1000 g of water, 300 g of talc particles having an average particle size of  $2.3\mu\text{m}$  and 165 g of 37 wt% formalin, followed by raising the temperature of the mixture and adding 38 g of phenol and 1.5 g of anhydrous sodium carbonate after the temperature reached  $85^\circ\text{C}$ . After the reaction was continued for one hour at that temperature, 113 g of melamine and 75 g of 10 wt% hydrochloric acid

were added and the reaction was further continued for additional 5 hours. Thereafter, sodium carbonate was added to the reaction mixture to neutralize it and to thus adjust pH thereof to 7. The reaction mixture was cooled to 50° C and was washed with 2 ℓ of water with filtering. The resultant solid contents were dried in a hot-air dryer maintained at 80 ° C until the weight of the solid became constant. The yield of the solid contents was 435 g. Thus, the talc particles covered with a layer of the co-condensate of formaldehyde, melamine and phenol, i.e., the composite particles of this invention were obtained. The composite particles had an average particle size of about 30 μ m.

#### Example 5

To a 4ℓ separable flask provided with a cooling apparatus, there were added 1000 g of water and 300 g of talc particles having an average particle size of 12 μ m to obtain a suspension. 2.5 g of 10 wt% hydrochloric acid was added to the suspension and the mixture was stirred at room temperature for 30 minutes. Then, 360 g of 37 wt% of formalin was added, the temperature of the mixture was raised up to 87 ° C and the mixture was maintained at that temperature for 40 minutes. To the mixture, there were added 180 g of melamine and 2.6 g of sodium hydroxide and the mixture was reacted at the same temperature for one hour with vigorously stirring. Further, 120 g of urea and 70 g of 10% hydrochloric acid and the reaction was continued at 60° C for additional 5 hours. Then, the contents of the flask was cooled down to 50 ° C and was washed with 2 ℓ of water with filtering. The resultant solid contents were dried in a hot-air dryer maintained

at 80 ° C until the weight of the solid became constant. The yield of the solid contents was 637 g. Thus, the talc particles covered with a layer of the co-condensate of formaldehyde, melamine and urea, i.e., the composite particles of this invention were obtained. The composite particles had an average particle size of about 15  $\mu$  m and the secondary particles were not observed therein.

#### Example 6

To a 4ℓ separable flask provided with a cooling apparatus, there were added 1200 g of water and 350 g of talc particles having an average particle size of 2.3  $\mu$  m to obtain a suspension of the talc particles. Then, 300 g of 37 wt% formalin and 240 g of melamine were added to the suspension, followed by raising the temperature thereof up to 90 ° C over 40 minutes and continuing the reaction at that temperature for 5 hours with vigorously stirring. In this respect, 3 g of a nonionic surfactant (NONION NS-210; available from Nippon Oils & Fats Co., Ltd.) was added 40 minutes after the temperature reached 90 ° C.

In addition, 0.1 N phosphoric acid solution was added for 30 minutes at a rate of 1 ml/min using a plunger pump 4 hours after the temperature reached 90° C.

Then, the contents of the flask were cooled down to 50° C and were washed with 2ℓ of water with filtering. The resultant solid contents were dried in a hot-air dryer maintained at 80 ° C until the weight of the solid became constant. The yield of the solid contents was 642 g. Thus, the talc particles which will be used in Example 7 were obtained. The composite particles had an average particle size



of about 4  $\mu$  m and the secondary particles were not observed therein.

#### Example 7

5 To a 4ℓ separable flask provided with a cooling apparatus, there were added 1800 g of 37 wt% formalin, 510 g of water, 1.8 g of 10 wt% aqueous solution of sodium hydroxide and 1590 g of melamine and these starting materials were reacted at 90 ° C for 60 minutes to thus obtain an initial condensation solution of melamine resin.

10 A third of the foregoing initial condensation solution and 634 g of the composite particles obtained in Example 6 were kneaded in a kneader and then was dehydrated under a reduced pressure. The solid thus obtained was dried in a hot-air dryer maintained at 80 ° C for 2 hours to obtain the amino resin composition of the present invention. The content of the talc particles in the composition was 27.5 wt%.

15 To the composition, there were added 0.4 part of zinc stearate and 0.1 part of phthalic anhydride per 100 parts of the composition and the mixture was pulverized in a ball mill to obtain a powdery molding material.

#### Comparative Example 1

20 A pulp-charged powdery molding material was prepared according to the same procedures used in Example 7 except that to a third of the initial condensation solution there was added 270 g of pulp in place of the composite particles.

#### Comparative Example 2

25 A talc-charged powdery molding material having a talc particle content of 27.5 wt% was prepared according to the same procedures used in Example 7 except that to a third of the initial condensation



solution there was added 241 g of talc particles having an average particle size of 2.3  $\mu$  m in place of the composite particles.

Mechanical properties of the molding materials obtained in Example 7 and Comparative Examples 1 to 2 are summarized in Table I given below.

Table I

Properties Measured	Ex. 7	Comp. Ex.1	Comp. Ex.2
Bending strength (kgf/mm <sup>2</sup> )	11.3	11.1	6.0
Bending modulus (kgf/mm <sup>2</sup> )	900	820	760
Charpy impact strength (kgf/cm <sup>2</sup> )	3.5	3.1	2.0

Then, the resistance to thermal discoloration was determined on the molded articles obtained from the molding materials prepared in Example 7 and Comparative Example 1. The relation between the degree of discoloration ( $\Delta E$ ) and the processing time is summarized in Table II given below.

The test conditions are as follows:

Processing temperature: 170° C

Atmosphere during the test: air

Colorimetric color difference meter used: Double Beam Cross-

Irradiation Photometric Z-1001DP Type available from Nippon

Denshoku Industries Co., Ltd.

Table II

Processing Time (hr.)	Degree of Discoloration ( $\Delta E$ )	
	Example 1	Comp. Example 1
0.5	2.0	10.2
1	3.1	14.4

2	4.6	18.7
4	6.4	24.8

As seen from the results listed in Table II, it is found that the molded article obtained from the molding material prepared in Example 7 exhibits mechanical strength identical to or higher than that observed on the molded article obtained using a cellulosic filler and has excellent resistance to thermal discoloration.

#### Example 8

To a kneader having a volume of 10ℓ, there were charged 925 g of talc particles having an average particle size of 2.3 μm, 1000 g of water, 2 g of 35% hydrochloric acid, 780 g of 37 wt% formalin and 75 g of pulp and the mixture was kneaded at room temperature for 10 minutes. 660 g of melamine was further added, followed by closing the kneader, raising the temperature up to 90° C with kneading and maintaining the mixture at that temperature for 80 minutes.

Then, the contents of the kneader were cooled to not more than 50 ° C and thus fluffy wet spherical particles having a particle size of 0.5 to 3 mm were obtained. The wet spherical particles were dried at 80 ° C till the weight of the particles became substantially constant to obtain the composite particles of this invention.

The composite particles (550 g) were mixed with 450 g of solid melamine resin free of fillers (IB-620; available from Reichhold Co., Ltd.) to obtain a composition. Then, to the composition there were added 0.4 part of zinc stearate and 0.1 part of phthalic anhydride per 100 parts of the composition, followed by pulverizing the mixture for 5 hours in a ball mill to obtain a powdery molding material.

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The mechanical properties of the foregoing molding material were determined and it was found that its bending strength was 10.0 kgf/mm<sup>2</sup>; that the bending modulus was 850 kgf/mm<sup>2</sup>; and that the charpy impact strength was 3.2 kgf/cm<sup>2</sup>. The molded article obtained from the molding material had a boiling water absorption as low as 0.2%. This means that the water resistance thereof is very good.

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What is claimed is:

1. Composite particles comprising talc particles covered with a layer of a condensate of at least one member selected from the group consisting of formaldehyde and formaldehyde-generating compounds and at least one member selected from the group consisting of aminotriazine type compounds, urea and phenols, the talc surface being chemically bonded to the condensate layer.
2. The composite particles of claim 1 wherein the content of talc ranges from 10 to 95% by weight on the basis of the total weight of the composite particles.
3. The composite particles of claim 1 wherein the particle size of the talc particles is not more than 60 mesh.
4. The composite particles of claim 1 wherein the aminotriazine type compound is selected from melamine, benzoguanamine and acetoguanamine.
5. The composite particles of claim 1 wherein the talc particles are replaced by pulp.
6. The composite particles of claim 1 wherein the phenol is selected from phenol, cresol, xylenol, t-butylphenol, phenylphenol, naphthol, alkyl phenols whose alkyl group has 2 to 12 carbon atoms, cyclohexylphenol, halogen-substituted phenols, resorcin, bisphenol A, bis(hydroxyphenyl)methane, bis(hydroxyphenyl) ether, bisphenol B and bisphenol S.
7. The composite particles of claim 1 wherein the formaldehyde-generating compound is selected from formalin, trioxane and paraformaldehyde.

8. The composite particles of claim 1 wherein they comprises secondary aggregates of the primary composite particles.
9. A method for preparing composite particles comprising talc particles covered with a layer of a condensate and in which the surface of the talc particles is chemically bonded to the condensate layer, which comprises the steps of: reacting at least one member selected from the group consisting of formaldehyde and formaldehyde-generating compounds formaldehyde with at least one member selected from the group consisting of aminotriazine type compounds, urea and phenols in an aqueous medium in the presence of talc particles to form the layer of the condensate on the surface of the talc particles.
10. The method of claim 9 wherein a desired amount of the talc particles is first added to the aqueous medium, then formaldehyde or the formaldehyde-generating compound is added to the resulting suspension and finally aminotriazine type compounds, urea and/or phenols are added.
11. The method of claim 9 wherein the talc particles are treated with an acid prior to the initiation of the condensation reaction.
12. The method of claim 9 wherein the aqueous medium is water and the amount thereof is selected so that the solid content at the end of the reaction ranges from 10 to 80% by weight.
13. The method of claim 9 wherein the reaction is carried out at a temperature ranging from 50 to 100 ° C for 1 to 10 hours.
14. The method of claim 9 wherein the molar ratio of formaldehyde or the formaldehyde-generating compound to aminotriazine type compounds, urea and/or phenols ranges from 1 to 3.

15. The method of claim 9 wherein an acid is added to the reaction mixture after the reaction proceeds to some extent.
16. An amino resin composition comprising the composite particles as set forth in claim 1.
17. The amino resin composition of claim 16 wherein the composite particles has a talc content ranging from 10 to 95% by weight based on the total weight of the composite particles.
18. The amino resin composition of claim 16 wherein the amount of the composite particles ranges from 5 to 70% by weight based on the total weight of the composition.
19. The amino resin composition of claim 16 wherein the amino resin is at least one member selected from the group consisting of melamine resins, urea resins, guanamine resins, phenol-melamine resins, guanamine-melamine resins, phenol-urea resins, urea-melamine resins, phenol-guanamine resins and urea-guanamine resins.
20. The amino resin composition of claim 16 wherein it further comprises a curing agent, a releasing agent, a coloring agent and/or other fillers.